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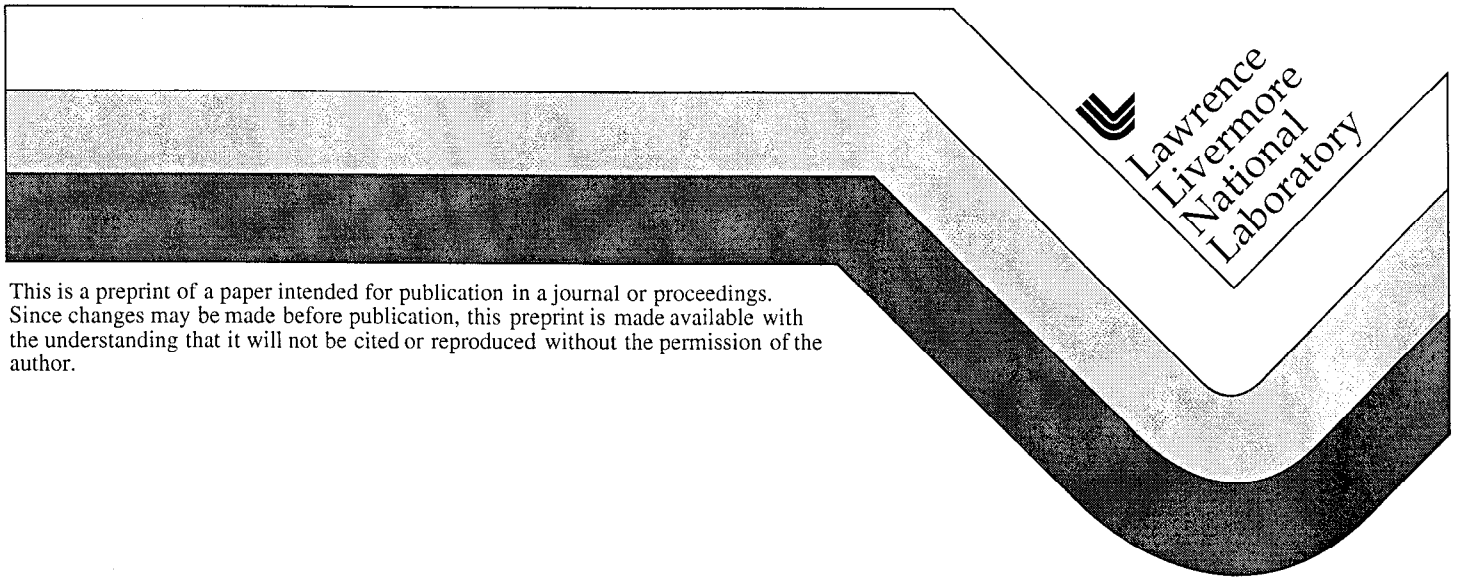

A Study of the Elasticity of Ta at High Temperature and Pressure

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The thermodynamic and transport properties of crystals and fluids at high temperature and pressure play a central role in the Earth and planetary sciences as well as in a variety of technologies and constitute a principal probe into the internuclear potential energy function at high density. Observation of surface waves by coherent time-domain optical spectroscopy provides an experimental approach to the determination, in the diamond-anvil high-pressure cell, of the elastic constants and thermal diffusivity of metal crystals as well as the equation of state and thermal transport properties of fluids. The electrical resistivities of metals may also be approximately accessible via the Wiedemann Franz Law. Preliminary results on Ta to pressures of 5.2 GPa are reported here. [Ta, elasticity, sound velocity, thermal diffusivity, equation of state]

I. Introduction

The elastic constants of an anisotropic single crystal are implicit in the velocity of surface acoustic waves as a function of crystallographic direction[1-2]. In all but a few metals, at pressure in excess of 1-2 GPa the velocity of longituidinal waves in many fluid pressure-transmitting media will exceed the speed of the surface wave in the metal. In this case the velocity of the interfacial wave is principally determined by the properties of the metal and the elastic constants of metals at high-pressure can be investigated by the coherent optical excitation and probing of interfacial waves (Stonely Waves) at the boundary between an oriented metal crystal and a pressure-transmitting medium. The velocities of interfacial waves are particularly sensitive to the shear moduli of the metal, these studies then complement the results of high pressure crystallography. Determination of the thermal diffusivity of the metal is concommittant of the measurements.

In homogeneous fluids that exhibit resonant optical absorption at a convenient laser wavelength, bulk compressional waves are excited and observed by thermally stimulated Brillouin spectroscopy[3-8]. The equation of state of the fluid can then be deduced from acoustic velocities. When the speed of sound in a fluid is less than that of Rayleigh Waves in the underlying metal, the velocity of the interfacial wave is largely determined by the properties of the liquid. The polished surface of a metal with a very high shear velocity (i.e. Be) or a metal mirror on a high-velocity substrate then constitutes a means of achieving, at high temperature and pressure, the signal strength characteristic of thermally stimulated Brillouin scattering in fluids such as the inert gases that do not absorb in the visible or near infrared.

2. Experimental Approach

The spectroscopic technique is discussed in general in Refs (3-5) and in the context of the diamond-anvil cell in (6-8). In outline, two "excitation" pulses, ≈80 ps in duration, selected from the output train of a Q-switched, mode-locked, Nd-YAG laser are combined at the sample surface at an angle 2θ, but otherwise coincident in space and time. Interference establishes a periodic distribution of intensity and the consequent temporally impulsive spatially periodic variation in the surface temperature launches a pair of counterpropagating surface acoustic waves of wavelength λ_A. The wavelength expressed in terms of λ, the wavelength of the laser light and θ is

$$d = \lambda_A = \frac{\lambda}{2\sin\theta} . \quad (1)$$

A third pulse selected from the same Q-switched envelope as the excitation pulses is doubled to 532 nm and delayed by time of flight to generate the "probe". Observation of the intensity of the Bragg scattering of the probe by the surface corrugation as a function of probe delay serves to determine the frequency (f_A), and hence the velocity ($c = \lambda_A f_A$), as well as the attenuation of the acoustic waves.

When the acoustic disturbance has been fully damped a spatially periodic distribution of temperature and hence surface corrugation remains. For sufficiently short wavelengths this "thermal grating" decays exponentially the rate constant, R, and time, τ, for the relaxation of the periodic distribution of temperature are given by

$$R = \frac{1}{\tau} = \frac{4\pi^2 D}{d^2} \quad (2)$$

where D is the thermal diffusivity (that is, the thermal conductivity divided by the heat capacity per unit volume). Since the scattered intensity is proportional to the square of the amplitude of the temperature grating, the directly observed decay rate is 2R. The variation of R with the angle of intersection, specified by Eqs.1 and 2 serves to distinguish one-dimensional thermal diffusion from other relaxation processes.

The simple form

$$I = \left(A_{th}e^{-Rt} - A_{ac}e^{-\gamma t}\cos\omega t\right)^2 \quad (3)$$

where A_{th} is the amplitude of the thermal grating, A_{ac} the amplitude of the acoustic grating, ω the circular acoustic frequency ($\omega = 2\pi f$), and γ the temporal acoustic attenuation coefficient gives a good account of the scattered intensity, I, as a function of probe delay t. In the ideal case of impulsive thermoacoustic excitation $A_{th} = A_{ac}$.

III. Representative Experimental Results

The velocities, determined by thermally stimulated Brillouin scattering, of Rayleigh waves propagating in the 100 plane of Ta at 25°C and 400°C are compared with those

calculated from elastic constants measured by conventional ultrasonic techniques in Fig. 1.

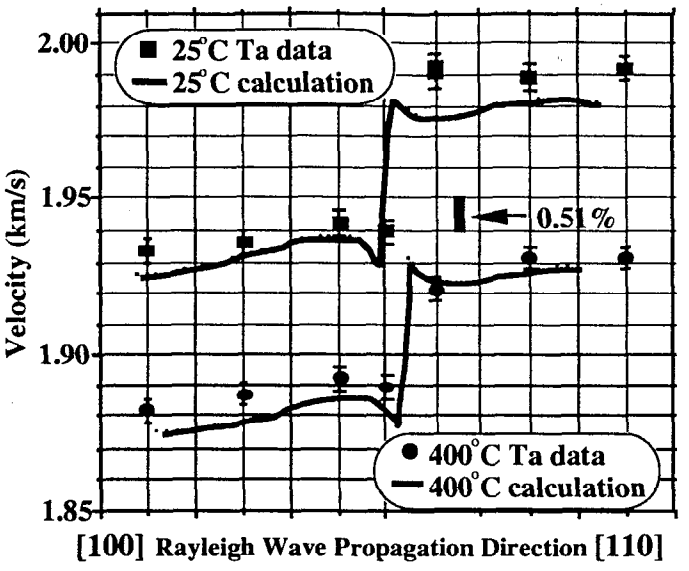

Fig. 1. The velocity of surface acoustic waves propagating in an approximate (100) plane of crystalline Ta at room temperature and at 400°C. The solid curves represent the velocities calculated from transducer based room-temperature measurements and presume a perfect (100) orientation.

Scattered intensity as a function of probe delay for the case of Ta at 25°C loaded with methanol in a diamond anvil cell at 5.2 GPa is given in Fig. 2. The Fourier transform of of this record, given in Fig. 3, corresponds to an acoustic frequency of 0.5632 GHz and a velocity of 2.064 km/sec. The signal to noise in the transform is seen to be excellent.

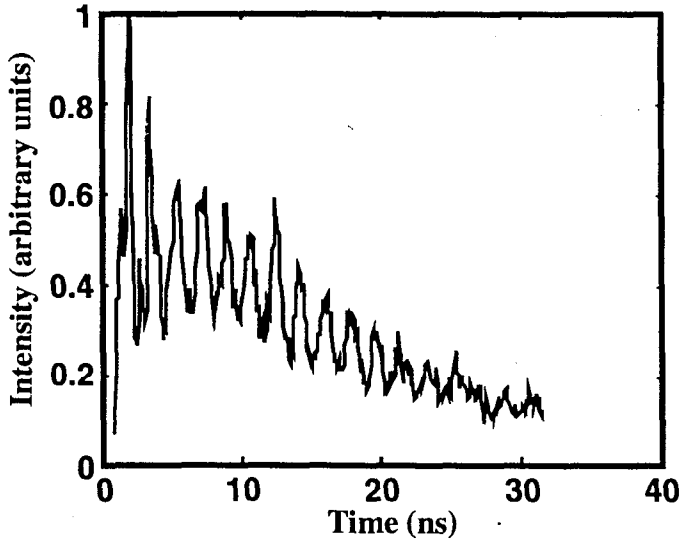

Figure 2. The scattered intensity as a function of probe delay for scattering by a Stonely wave with a wavelength of 3.82 μm at a Ta - methanol interface in a diamond anvil cell at 25°C and 5.2 GPa.

III. Summary

These experiments demonstrate that the observation of the time evolution of thermoacoustically excited surface waves is a feasible route to the determination of the elastic moduli of metals at high temperature and pressure. Experiments on the

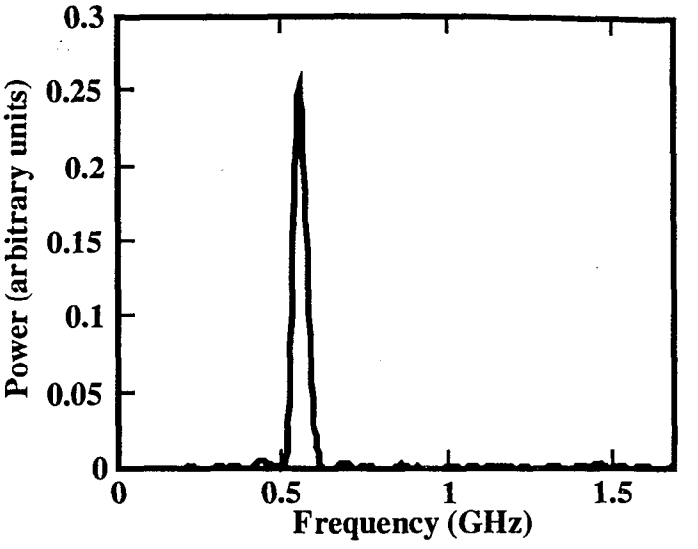

Figure 3. The Fourier transform of the record in Fig. 2. The peak frequency is 0.5632 GHz.

velocity of Stonely waves as a function of crystallographic direction, pressure, and temperature in tantalum single crystals are underway. The signal to noise ratio associated with observations of scattering from mirror-like samples in the diamond-anvil cell is determined principally by laser fluctuations and scattering from particulate contaminates on the various surfaces. It is reasonable to expect that the results in Fig. 3 can be achieved for any smooth metal surface.

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